AMS ^{14}C DATES AND MAJOR ELEMENT COMPOSITION OF GLASS SHARDS OF LATE PLEISTOCENE TEPHRAS ON TANEGASHIMA ISLAND, SOUTHERN JAPAN

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ABSTRACT. Four late Pleistocene tephra layers—Tane I (Tn1), II (Tn2), III (Tn3), and IV (Tn4) in ascending order—are intercalated between widespread tephras, Kikai-Tozurahara (K-Tz: 95 ka) and Aira-Tn (AT: 30 cal kBP), on Tanegashima Island, in southern Japan. Paleolithic ruins such as the Yokomine C and Tatikiri archaeological sites were excavated from the loam layer between the Tn4 and Tn3 tephras. To refine the chronological framework on the island, we conducted accelerator mass spectrometry (AMS) radiocarbon dating for 2 paleosol and 6 charcoal samples related with the late Pleistocene tephras and the Yokomine C archaeological site. The obtained ^{14}C dates are consistent with the stratigraphy in calendar years, 33 cal kBP for Tn4, 40 cal kBP for Tn3, and >50 cal kBP for Tn2 and Tn1. The charcoal dates from Yokomine C, 32–38 cal kBP, not only constrain the age of Tn4 and Tn3 ashes, but also serve as a possible date for the site. We also measured the major element compositions of volcanic glass shards with EDS-EPMA to characterize these tephras. Although we could not find a possible correlative for Tn3 and Tn4 ashes using major element oxides of the glass shards, i.e. 75–76 wt% in SiO_2, the glass chemistry obtained in this study will be valuable in correlating these tephras with their source volcanoes in the near future.

INTRODUCTION

Tanegashima Island is located 20 km off the south coast of Kyushu Island (Figure 1). Four late Pleistocene tephras—Tane I (Tn1), II (Tn2), III (Tn3), and IV (Tn4) in ascending order—are intercalated between 2 widespread tephras, Kikai-Tozurahara (K-Tz: Nagaoka 1988; Machida and Arai 1992) and Aira-Tn (AT: Machida and Arai 1976), on the island (Figure 2). Widespread tephras provide useful time-markers for various related studies (Machida and Arai 2003). Paleolithic ruins such as the Tatikiri (Nakatane Town Board of Education 2003) and Yokomine C (Minamitane Town Board of Education 2000) archaeological sites are located in the southern part of Tanegashima Island (Loc. 4 and 6 in Figure 1B), and excavated from the loam layer between the Tn3 and Tn4 ashes. The radiocarbon dates of charcoal fragments reported previously ranged from 28.4 to 30.4 kBP and 29.6 to 31.3 kBP for the Tatikiri and Yokomine C sites, respectively (Table 1). These dates provide constraints for estimating the eruption ages of the Tn3 and Tn4 ashes.

Since the discovery of the AT ash (Machida and Arai 1976), the catalog of tephras in and around the Japanese Islands has been compiled and subsequently revised (e.g. Machida 2002; Machida and Arai 1992, 2003). The Tn1 and Tn2 tephras can be correlated with the Kiyomidake (Ata-Ky) tephra that erupted from Ata caldera (Figure 1B; Okuno et al. 1995) on the basis of mineral assemblages and refractive index of opx (γ). However, the source volcanoes for the Tn3 and Tn4 ashes have not been identified yet (Okuno and Kobayashi 1994). Recently, Moriwaki et al. (2009) obtained the major chemical compositions of glass shards from late Pleistocene and Holocene tephra layers in the Tokara Islands (Figure 1A) using a wavelength dispersive electron probe microanalyzer (WDS-EPMA). Knowing the glass chemistry enables us to find the distal correlatives.

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Proceedings of the 6th International Radiocarbon and Archaeology Symposium, edited by E Boaretto and N R Rebollo Franco
RADIOCARBON, Vol 54, Nr 3–4, 2012, p 351–358

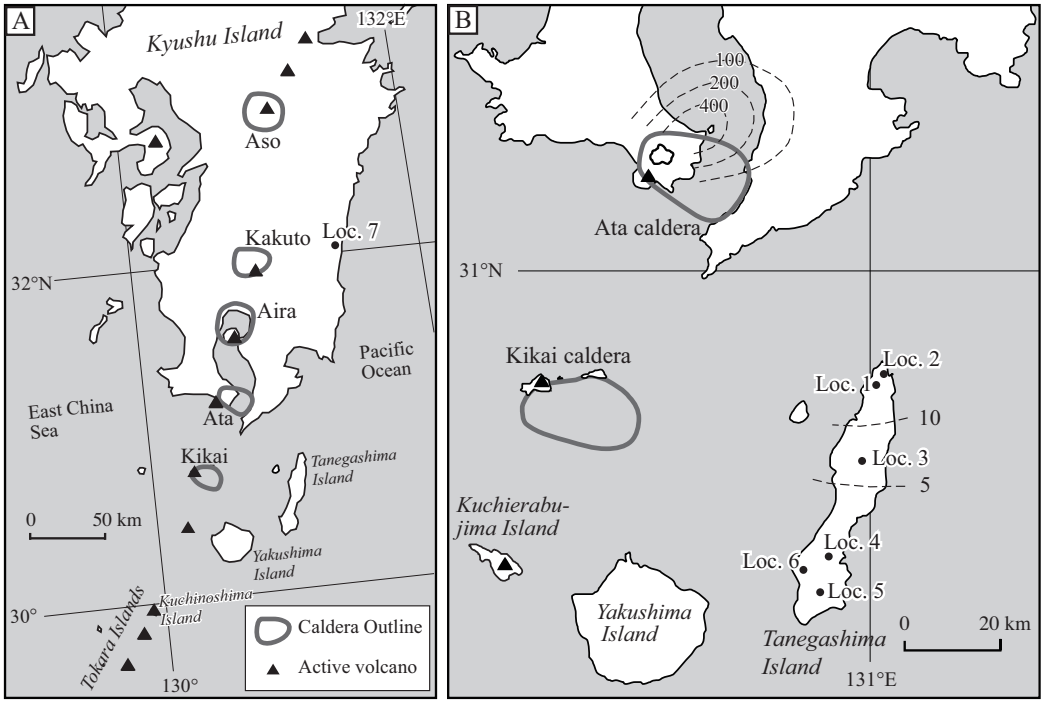

Figure 1 Index maps: A) Map showing location of Tanegashima Island; B) Dashed lines show isopachs of Tn2 pumice on Tanegashima Island (Okuno and Kobayashi 1994) and Ata-Ky tephra around Ata caldera (Okuno et al. 1995), respectively, with values given in cm. Solid circles with numbers on Tanegashima Island show the localities of sampling and archaeological sites: Loc. 1: Kunigami Junior High School, Nishino-omote City (30°47′44″N, 131°02′14″E); Loc. 2: Kubota, Nishino-omote City (30°49′19″N, 131°03′48″E); Loc. 3: Furuta, Nishino-omote City (30°40′10″N, 131°00′47″E); Loc. 4: Tatikiri archaeological site, Nakatane Town (30°27′59″N, 130°56′15″E); Loc. 5: Nakanoshita, Minamitane Town (30°23′50″N, 130°54′01″E); Loc. 6: Yokomine C archaeological site, Minamitane Town (30°26′40″N, 130°52′24″E); Loc. 7: Nyutabaru, Shintomi Town (32°05′21″N, 131°27′39″E).

Table 1 Previously reported AMS radiocarbon dates for the Yokomine C (Minamitane Town Board of Education 2000) and Tatikiri (Nakatane Town Board of Education 2003) archaeological sites.

Locality	$\delta^{13}C_{PDB}$ (‰)	^{14}C age (BP)	Lab code (Beta-)	Calibrated date (cal BP, 2σ)
Tatikiri archaeological site (Loc. 4)				
	–24.3	30,400 ± 600	169709	33,653–36,386 (100%)
	–26.4	28,400 ± 500	169710	31,505–34,162 (99.1%) 34,341–34,417 (0.9%)
Yokomine C archaeological site (Loc. 6)				
	–25.8	31,280 ± 690	102399	34,591–37,152 (100%)
	–24.6	29,670 ± 540	102400	32,899–35,185 (100%)
	–24.3	30,490 ± 590	102401	33,861–36,469 (100%)

To refine the chronological framework on Tanegashima Island, we conducted ^{14}C dating of paleosol and charcoal samples using accelerator mass spectrometry (AMS) techniques. We also analyzed the major chemical compositions of volcanic glass shards using an energy dispersive electron probe microanalyzer (EDS-EPMA) to characterize these tephra layers.

STRATIGRAPHY OF THE LATE PLEISTOCENE TEPHRAS ON TANEGASHIMA ISLAND

Representative columnar sections on the island are shown in Figure 2. According to Okuno and Kobayashi (1994), Tn1, an orange fine ash, and the overlying Tn2 pumice are distributed on the northern part of the island (Figure 1B). On the other hand, the 2 upper fine yellowish ash layers, Tn3 and Tn4, cover the whole area of the island without a systematic change in thickness. All tephras contain plagioclase, orthopyroxene, and clinopyroxene as phenocryst. A small amount of hornblende and quartz phenocrysts is contained in the Tn3 and Tn4 ashes.

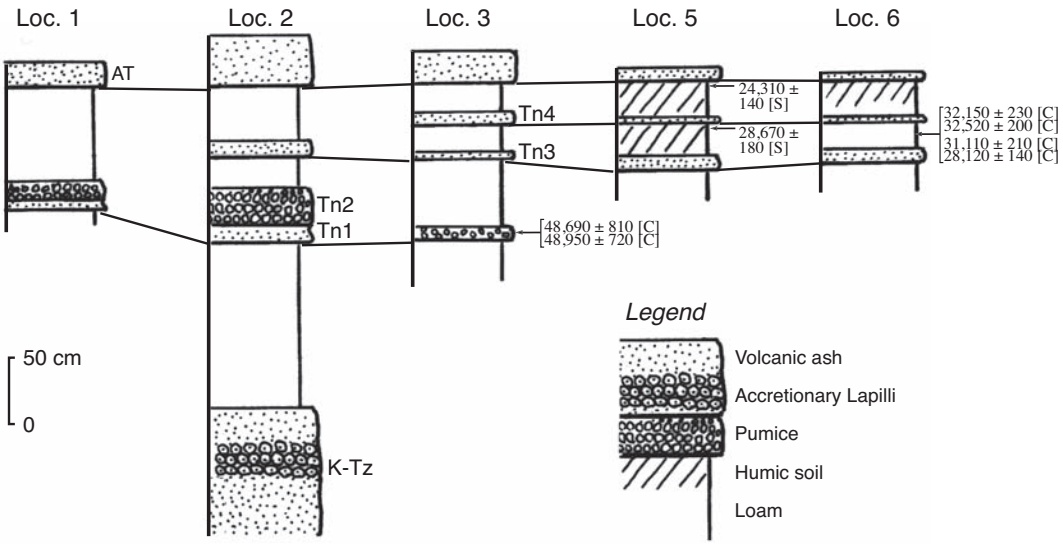

Figure 2 Columnar sections of Late Pleistocene tephras on Tanegashima Island. The location sites are shown in Figure 1. Solid arrowheads indicate horizons of tephra samples.

SAMPLES AND ANALYTICAL METHODS

For AMS ^{14}C dating, we collected 2 paleosol samples 2 cm thick immediately below the tephra layers AT and Tn4 and 3 charcoal samples, 2 between Tn4 and Tn3 and 1 within Tn2 (Figures 1B and 2; Table 2). For the EDS-EPMA analyses of glass shards, we also collected samples from each tephra layer (Figures 1B and 2; Table 3). In addition, we collected AT ash from Loc. 7 (Figure 1A) on Kyushu Island, to check the accuracy and precision of EDS-EPMA analyses. The sampling horizon was 5 cm above the bottom of the AT-II unit (Nagaoka 1988).

AMS Radiocarbon Dating

Paleosol and charcoal samples were cleaned chemically with acid-alkali-acid (AAA) treatment and combusted to CO_2. The soil sample was washed and dispersed in distilled water using an ultrasonic cleaner before AAA treatment. The samples were then wet-sieved using a 106-μm sieve to remove contaminating materials (Okuno et al. 1997, 2001). The CO_2 produced was cryogenically purified

Table 2 Results of AMS radiocarbon dating.[a]

Locality	Stratigraphic position	Material	$\delta^{13}C_{PDB}$ (‰)	^{14}C age (BP)	Lab code (JAT-)	Calibrated date (cal BP, 2σ)
Loc. 5	Below AT	Soil	–15.2	24,310 ± 140	7816	28,598–29,493 (100%)
	Below Tn4	Soil	–16.2	28,670 ± 180	7815	32,246–33,678 (97.5%)
						33,757–34,001 (2.5%)
Loc. 6	Between Tn4 and Tn3	Charcoal	–27.1	32,150 ± 230	7817	35,727–35,812 (1.0%)
						36,268–37,320 (99.0%)
		Charcoal	–27.5	32,520 ± 200	8702	36,529–37,683 (100%)
		Charcoal	–24.7	31,110 ± 210	8668	35,047–36,322 (100%)
		Charcoal	–28.7	28,120 ± 140	8701	31,706–32,926 (100%)
Loc. 3	In Tane 2	Charcoal	–22.1	48,690 ± 810	8667	
		Charcoal	–29.1	47,950 ± 720	8700	

[a]Note: Sampling sites are shown in Figure 1.

and then converted catalytically to graphite (Kitagawa et al. 1993) and was measured by the JAEA-AMS-TONO at the Tono Geoscience Center, JAEA (Xu et al. 2000). All 3 carbon isotopes were measured with the AMS in both the samples and the NIST oxalic acid (HOxII) standard. To estimate the ^{14}C background level, the ^{14}C content of IAEA C1 (marble) was also measured in the same sequence of sample measurements. The ^{14}C age was calculated by subtracting the ^{14}C concentration of the background sample. We corrected for carbon isotopic fractionation using the $^{13}C/^{12}C$ ratio ($\delta^{13}C_{PDB}$) to calculate the conventional ^{14}C ages. The ^{14}C errors were evaluated by ^{14}C reproducibility of repeated measurements on standard targets. Conventional ^{14}C dates were calibrated to the calendar year timescale using the IntCal09 data set (Reimer et al. 2009) and CALIB 6.0 program (Stuiver and Reimer 1993; www.calib.org).

EDS-EPMA Analysis

Tephra samples were washed using an ultrasonic cleaner and wet-sieved using a 1/16-mm sieve. The remaining particles were dried at <40 °C and embedded in mounts made of polyester resin. Mount surfaces were polished with diamond paste (1 μm) until the internal surfaces of the glass particles were exposed, and were then coated with carbon. We analyzed the major oxide compositions of 9 major elements (Si, Ti, Al, Fe, Mn, Mg, Ca, Na, and K) in the volcanic glass shards using a scanning electron microscope (SEM; JEOL JSM-7001F) equipped with an energy dispersive X-ray spectrometer (EDS; Oxford INCA X-Max) at the Faculty of Science, Kumamoto University, Japan. The EDS accelerating voltage was 15 kV, the specimen current was 1 nA, the live time was 50 s, and the beam scanning area was 10 μm wide for all samples apart from the Tn1 tephra. We could not find a scanning area of 10 μm wide for the Tn1 tephra. To avoid the loss of alkaline components during measurement, we have to change the measurement conditions, to a specimen current of 0.6 nA and a beam scanning area 2 μm wide. We used the ZAF correction method (Sweatman and Long 1969) to calculate the oxide abundances. We measured 1 point on each volcanic glass shard and 10–20 points per sample. The accuracy and precision of this analysis were checked by an obsidian standard (No.33 obsidian, Astimex Scientific Ltd.). The AT ash was used as an in-house standard because it is very homogeneous with respect to the major elements (e.g. Machida and Arai 2003; Moriwaki et al. 2011).

RESULTS AND DISCUSSION

Radiocarbon Dates

The ^{14}C dates obtained for soil samples below the AT and Tn4 ashes were 24,310 ± 140 and 28,670 ± 180 BP, respectively. Dates of the charcoal fragments between the Tn4 and Tn3 ashes at Loc. 6 are 28,080 ± 130, 31,160 ± 250, 32,150 ± 230, and 32,580 ± 180 BP, and those in the Tn2 pumice deposit are 47,950 ± 720 and 48,690 ± 810 BP, respectively (Table 2). All ^{14}C dates are consistent with the stratigraphy (Figure 2), previous ^{14}C dates for the AT tephra (~24–25 kBP; e.g. Ikeda et al. 1995; Miyairi et al. 2004), and for the archaeological sites (28–32 kBP; Table 1). The ^{14}C date of a soil sample just below a tephra represents the time when the tephra covered the surface soil, thus giving its eruption age (Okuno et al. 1997, 2001; Okuno and Nakamura 2003). Hence, the age of the Tn4 ash is estimated to be 33 cal kBP. The charcoal dates from Loc. 6 (32–38 cal kBP) are similar to the age of the Tn4 ash. The calibrated calendar years for the Tn2 pumice exceed the range of calibration data set of IntCal09 (~50 cal kBP).

The eruption age can be estimated tentatively by assuming a constant accumulation rate for the loam layers between time-marker tephras (e.g. Hayakawa 1991, 1995). Based on the ages of K-Tz (95 ka; Machida and Arai 2003) and AT (30 cal kBP; Okuno et al. 2011), the ages of the late Pleistocene tephras in Tanegashima Island are estimated to be 67 ka for Tn1 and Tn2, 46 ka for Tn3, and 39 ka for Tn4. On the other hand, the age of Ata-Ky in the Osumi Peninsula is also estimated to be 57 ka, which is more consistent with the ^{14}C dates of the charcoal fragments in the Tn2 pumice in this study. This inconsistency in eruption age between the 2 areas implies that the assumption of constant accumulation of loam is not strictly applicable. However, the ^{14}C dates from the archaeological sites and the stratigraphic relationship with Tn4 and Tn2 enable us to estimate the age of the Tn3 to be approximately 40 cal kBP.

Major Chemical Compositions of Volcanic Glass Shards

Table 3 shows the mean values and standard deviations of major chemical compositions of volcanic glass shards in each sample. All shards were vitreous and isotropic and showed no sign of alteration in transmitted or reflected light. The major chemical compositions of the obsidian standard were in good agreement with the recommended value. The major chemical compositions of the AT volcanic glasses were very homogeneous (Table 3) and consistent with those reported by previous studies (e.g. Machida and Arai 2003; Moriwaki et al. 2011).

The variation diagrams of major element oxides vs. SiO_2 (Figure 3) shows that the Tn3 and Tn4 ashes have similar chemical composition, i.e. 75–76 wt% in SiO_2 at different sites (Loc. 5 and 6). Glass of the Kc-5 tephra that erupted from Kuchinoshima Volcano (Moriwaki et al. 2009), which could be placed in a similar horizon, is significantly higher in SiO_2 than the Tn3 and Tn4 ashes. The N-Ym tephra that erupted from Kuchierabujima Volcano (Moriwaki et al. 2009) has a similar trend as the Tn3 and Tn4 ashes in Figure 3, but with a stratigraphic position above the AT ash. Although we cannot report yet the possible correlative for the Tn3 and Tn4 ashes, the chemical characteristics reported in this study will enable us to identify the source vents in the near future. On the other hand, the major element oxides of the volcanic glass from the Tn2 pumice and the Tn1 ash were of similar value in SiO_2 (~74 wt%). This resemblance in glass chemistry indicates that both tephras were successively erupted from the same volcano.

Table 3 Major element compositions in weight % (wt%; normalized to an anhydrous basis) of glass shards of the late Pleistocene tephras on Tanegashima Island.

Tephra name	Locality		SiO_2	TiO_2	Al_2O_3	FeO[a]	MnO	MgO	CaO	Na_2O	K_2O	Cl	Total	*n*	Remarks
Tn1	Loc. 2	Av[b]	74.11	0.57	12.78	3.04	0.13	0.51	2.23	3.29	3.36		97.46	2	
		SD	0.06	0.01	0.28	0.04	0.15	0.12	0.39	0.43	0.74		0.88		
Tn2	Loc. 1	AV.	74.22	0.60	12.56	3.03	0.07	0.57	2.35	3.69	2.89		98.23	20	
		SD	0.25	0.08	0.09	0.24	0.07	0.06	0.11	0.11	0.16		1.31		
Tn3	Loc. 5	AV.	75.44	0.60	12.29	2.38	0.11	0.40	1.75	3.96	3.08		98.50	21	
		SD	0.40	0.05	0.20	0.13	0.11	0.05	0.14	0.09	0.10		0.84		
	Loc. 6	AV.	75.80	0.60	12.12	2.32	0.08	0.39	1.66	3.95	3.09		98.05	16	
		SD	0.31	0.09	0.12	0.11	0.08	0.04	0.11	0.13	0.10		0.74		
Tn4	Loc. 5	AV.	75.37	0.58	12.31	2.46	0.08	0.42	1.82	3.95	3.01		98.77	20	
		SD	0.29	0.07	0.15	0.18	0.08	0.05	0.14	0.14	0.19		0.90		
	Loc. 6	AV.	75.44	0.60	12.36	2.35	0.07	0.44	1.86	3.92	2.96		98.43	15	
		SD	0.32	0.09	0.12	0.15	0.07	0.05	0.11	0.13	0.06		0.52		
AT	Loc. 7	AV.	77.63	0.15	12.28	1.28	0.06	0.11	1.19	3.78	3.52		94.18	20	AT-II
		SD	0.26	0.08	0.12	0.13	0.07	0.05	0.07	0.09	0.12		1.70		
Obsidian		AV.	74.55	0.07	13.05	1.57	0.10	0.03	0.81	4.05	5.42	0.35	97.62	17	Astimex Scientific Ltd.
		SD	0.16	0.07	0.09	0.11	0.07	0.04	0.05	0.08	0.08	0.03	0.62		
Sz-S	Satsuma Peninsula	AV.	74.58	0.39	13.48	1.94	0.05	0.52	2.15	3.96	6.53		93.44	29	Moriwaki et al. 2011
		SD	0.48	0.09	0.35	0.11	0.03	0.16	0.24	0.12	0.13		1.83		
N-Ym Upper	Kuchierabujima	AV.	77.37	0.34	12.43	1.42	0.06	0.31	1.49	3.67	2.91		95.50	9	Moriwaki et al. 2009
		SD	1.84	0.17	0.47	0.61	0.06	0.24	0.67	0.12	0.19		1.90		
N-Ym Middle	Kuchierabujima	AV.	73.29	0.61	13.27	3.27	0.10	0.64	2.72	3.38	2.71		91.78	11	Moriwaki et al. 2009
		SD	0.87	0.09	0.48	0.33	0.04	0.16	0.43	0.18	0.19		3.67		
N-Ym Lower	Kuchierabujima	AV.	74.90	0.49	12.94	2.47	0.06	0.48	2.14	3.61	2.88		94.28	10	Moriwaki et al. 2009
		SD	0.42	0.09	0.17	0.18	0.05	0.05	0.11	0.17	0.06		1.38		
K-Ah	Nakanoshima	AV.	74.74	0.54	12.97	2.58	0.07	0.46	2.06	3.91	2.68		97.36	10	Moriwaki et al. 2009
		SD	0.54	0.16	0.16	0.22	0.04	0.08	0.18	0.15	0.13		0.71		

[a]Note: the original analytical total is also presented.

[b]Av = average value; SD = standard deviation

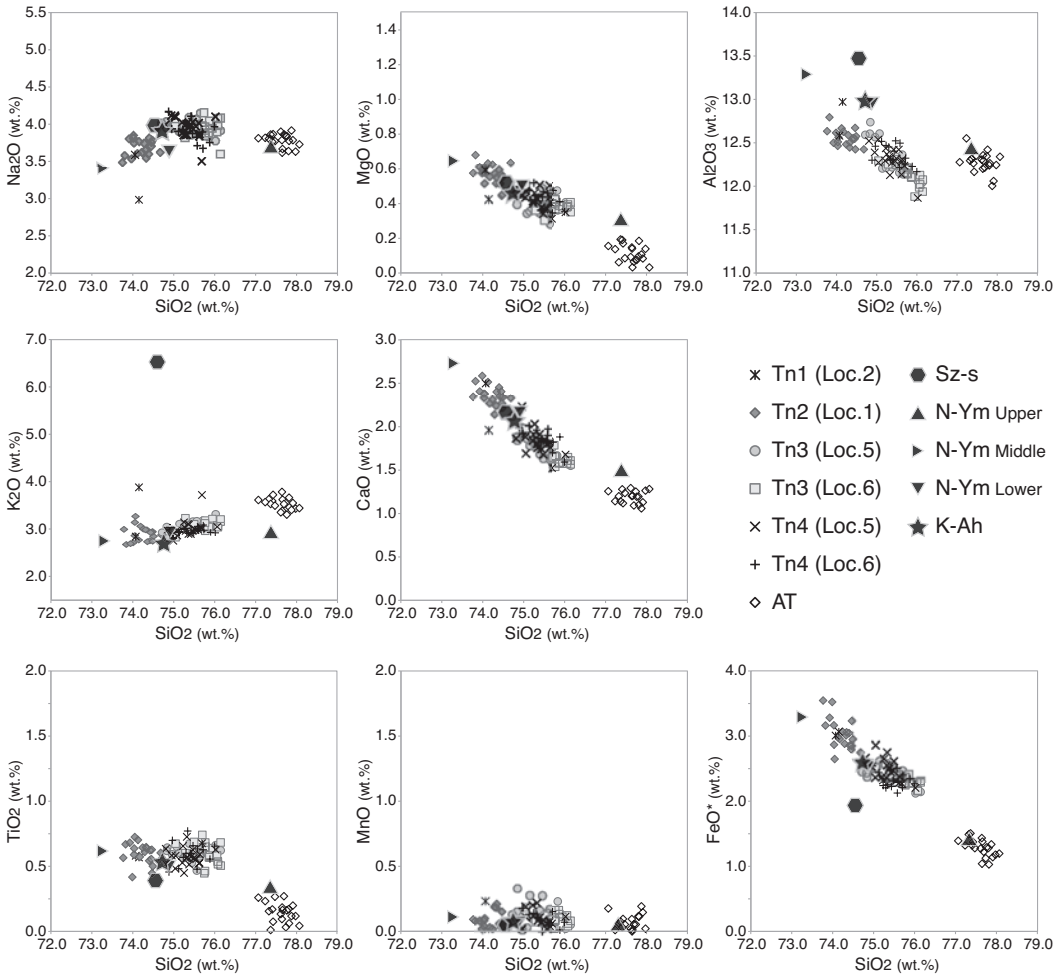

Figure 3 Harker variation diagrams in weight % (wt%; normalized to an anhydrous basis) for glass shards of Late Pleistocene tephras on Tanegashima Island. Selected tephras from Tokara Islands (Moriwaki et al. 2009) and Sakurajima Volcano (Moriwaki et al. 2011) are also plotted for comparison. * = Total iron as FeO.

CONCLUSION

The obtained ^{14}C dates for the late Pleistocene tephra layers were mostly consistent with the stratigraphy and previously reported ^{14}C dates. The eruption ages of these tephras were estimated as 33 cal kBP for Tn4 ash, 40 cal kBP for Tn3, and >50 cal kBP for Tn1 and Tn2. We could not find a possible correlative for the Tn3 and Tn4 ashes using major element oxides of the glass shards. Further studies are needed until we can use the glass chemistry of these tephras to identify the volcanic source.

ACKNOWLEDGMENTS

This study was financially supported by a Grant-in-Aid for Scientific Research (Nos. 17300295, 21101002) from the Japan Society for the Promotion of Science (JSPS) and the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan, and Sasakawa Grants for Science Fellows from the Japan Science Society. AMS ^{14}C dating in this study was performed under the Common-Use Facility Program of JAEA. We thank Prof Tadao Nishiyama and Associate Prof Hiro-

shi Isobe of Kumamoto University for their support of the EDS-EPMA analysis. We are grateful to Dr Ma Hannah T Mirabueno (Philippine Institute of Volcanology and Seismology) for constructive comments that improved the manuscript. We dedicate this paper to the late Professor Shinji Nagaoka of Nagasaki University for his contributions in tephrochronology in southern Kyushu.

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