# Eruption Age of Sakurajima-Satsuma Tephra Using Thermoluminescence Dating

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#### 1. Introduction

Sakurajima, located on the southern volcanic front in Kyushu, is a very active volcano in Japan. Tephra beds from Sakurajima volcano are collectively designed as the Sakurajima-tephra (Sz) group (Kobayashi, 1986). The eruption history of Sakurajima volcano is classified tephrostratigraphically into the following three stages: Old Kitadake, Young Kitadake, and Minamidake (Kobayashi and Ezaki, 1997). The first eruption of the Young Kitadake stage is designated as Sakurajima-Satsuma (Sz-S or P14) tephra (Kobayashi and Ezaki, 1997), which is volcanic ash of the largest eruption of the Sz group (e.g. Moriwaki, 1992). Sz-S derived from Sakurajima volcano is a key marker for volcanic stratigraphy, palaeoenvironmental studies (e.g. Moriwaki, 1992), and archaeological studies (e.g. Kodama, 2001) in

southern Kyushu and surrounding marine regions of Sakurajima volcano because this tephra, with the largest volume (ca. 11 km³: Kobayashi and Tameike, 2002) of Sakurajima tephras, is widespread throughout southern Kyushu. Therefore, age determination of this key tephra layer (Sz-S) is expected to contribute greatly to studies in many fields.

Regarding the ages of Sz-S, Machida and Arai (1992) reported a radiocarbon (<sup>14</sup>C) age of 10,500 BP as an average of several charcoal ages obtained using the beta counting method. Okuno (1997) obtained accelerator mass spectrometry (AMS) <sup>14</sup>C ages of charcoal samples from tephra beds of Sz group, and soil materials related closely to the tephra. The reported <sup>14</sup>C ages are presented in Table 1. Okuno (2002) estimated it as 12,800 cal yrBP based on the <sup>14</sup>C dates shown in Table 1.

Table 1	Reported 14C ag	es of samples	related to Sz-S	(Okuno 1997)
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Stratigraphic horizon	Material	<sup>14</sup> C date (yrBP)	Calibrated age (cal yrBP)	Lab no.
In Sz-S	Charcoal	10,670±100	12,700-12,400	NUTA-4634
In Sz-S	Charcoal	11,050±120	13,100-12,700	NUTA-4642
Below Sz-S	Soil	10.910± 80	12,990–12,690	NUTA-3784
Below Sz-S	Soil	11,280± 80	13,300-13,000	NUTA-3878
Below Sz-S	Soil	11,330± 90	13,370-13,050	NUTA-4025
Below Sz-S	Soil	11,660±100	13,700–13,300	NUTA-3868
Below Sz-S	Soil	11,850± 90	13,940–13,460	NUTA-3561

Calibrated ages were estimated using the IntCal13 data set (Reimer et al., 2013)

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Because of charcoal that is present in or below soil, the possibility of contamination of the volcanic ash layer by old charcoal or a humic fraction from exotic materials cannot be eliminated completely. For such samples, <sup>14</sup>C dating sometimes misrepresents the true age. Therefore, other dating methods are useful to confirm the <sup>14</sup>C age. Thermoluminescence (TL) dating is a candidate method for such an investigation because the TL age, obtained from the tephra itself, is the age of the latest heating.

The TL dating of quartz has clarified the history of young (quartz-bearing) silicic volcanisms (e.g. Gillot et al., 1978; Guerin and Valladas, 1980; Ichikawa et al., 1982). Recently, Toyoda et al. (2006) clarified much of Japanese Quaternary silicic volcanisms by dating of Isothermal-TL and ESR ages of quartz in several tephra layers. Ganzawa et al. (2007) estimated the eruption age of Toya pyroclastic flow as 113-114 ka using the single aliquot regenerative-dose (SAR) protocol for quartz analysis (Murray and Wintle, 2000). The TL dating of feldspar is a candidate for age determination of young quartz-less volcanic rocks (Aitken, 1985). However, the applicability of TL dating of feldspar remains controversial because the feldspar readily causes anomalous fading of luminescence signals (Wintle, 1973).

Shitaoka *et al.* (2009a) reported that polymineral fine grains (PFG) have great potential for use as targets of TL dating of quartz-less volcanic rocks. Shitaoka *et al.* (2009a) reported that TL age obtained from PFG analysis, within error, to that obtained from quartz analysis. Consequently, TL dating of PFG in quartz-less tephra is a powerful tool for establishment of details of the young volcanic stratigraphy (e.g. Shitaoka *et al.*, 2009b; Oishi *et al.*, 2011; Buvit *et al.*, 2014).

Palaeodoses estimations have used traditionally multiple-aliquot protocol (Aitken, 1985) or mainly single-aliquot protocol (Murray and Wintle, 2000). The tephra sample deposited near the source vent collected from a thick and less-altered layer provides the most probable age of the tephra because it is a nearly closed system (Shitaoka and Nagatomo, 2011). In this case, it is reasonable to use the multiple aliquot additive dose (MAAD) protocol (Aitken, 1985) because measure-

ments of SAR take much more time than measurements of the MAAD protocol.

The Sz-S tephra does not contain the quartz. Therefore, we addressed age determination of the sample using the TL method of PFG with the MAAD protocol.

#### 2. Stratigraphic positions of tephra samples

We collected samples of the Sz-S tephra bed from two locations (Fig. 1) on the Osumi (Loc. 1) and Satsuma Peninsulas (Loc. 2). Loc. 1 is on the Aira caldera wall at ca. 12 km east of Sakurajima volcano. Loc. 2 is at an archaeological site in the campus of the Faculty of Medicine, Kagoshima University, ca. 13 km southwest of the volcano.

At Loc. 1, a sequence of most Sakurajima marker tephras and two tephras from other volcanoes overlie the Ito pyroclastic flow deposit (Aramaki, 1984). The five tephras were identified as Sz-Sy (P11), Sz-Ub (P12), Sz-Tk3 (P13), Sz-S and Takano base surge (Tn-bs) from the upper to lower (Kobayashi and Fukushima, 1984; Moriwaki, 1994). Of these tephras in this section, Sz-S tephra is identified as a distinct tephra occurring stratigraphically in the middle part of the sequence based on several distinct units including a fine-grained phreatomagmatic ash fall, in contrast to

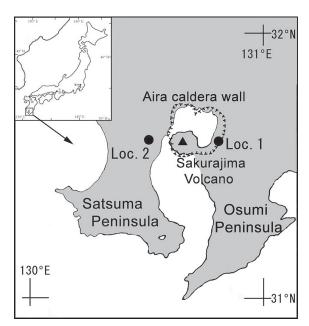


Fig. 1 Locality map of sampling points: Loc. 1 is 31° 35' 18"N, 130° 47' 35"E: Loc. 2 is 31° 32' 53"N, 131° 31' 45"E.

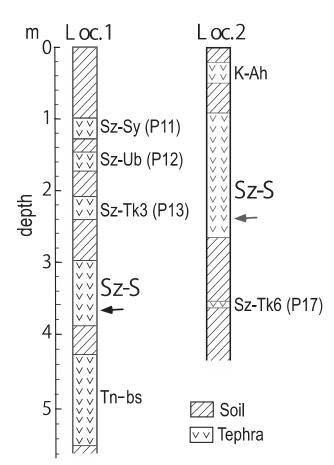


Fig. 2 Geologic logs around Sz-S tephra and TL samples at Loc. 1 and Loc. 2. Sz-Sy (P11), Sz-Ub (P12), Sz-Tk 6 (P17) and Tn-bs (Kobayashi and Ezaki, 1997) are Sz group. Arrows indicate sampling horizons.

other tephras (Kobayashi, 1986; Moriwaki, 1992, 1994) (Fig. 2, Loc. 1). This tephra overlies the Tn-bs deposit (Kobayashi and Ezaki, 1997) derived from the Aira caldera, of which the facies differ distinctly from those of other fall tephras. The Sz-S tephra, which is 70 cm thick, consists mostly of pumice grains (Fig. 2, Loc. 1). The sample analyzed in this study was collected from the lowermost coarse pumice fall unit (Fig. 2, Loc. 1).

At Loc. 2, three marker tephras overlie the Ito pyroclastic flow deposit (Fig. 2, Loc. 2). Each tephra bed shows distinct lithofacies: the upper consisting of a finegrained vitric ash bed, the middle one with many tephra units, and the lower one consisting of brown pumice with bluish accidental lithics. Based on these features, together with stratigraphic positions, the three tephras were identified as Kikai-Akahoya tephra (K-Ah; Machida and Arai, 1992), Sz-S, and Sz-Tk6 (P17) from the upper to lower (Kobayashi and Fukushima, 1984; Moriwaki, 1994). Of the more than 13 units

for Sz-S (Moriwaki, 1994), more than 6 units occur in this section. A base surge deposit and two fine-grained ash falls are intercalated with pumice falls. An analyzed sample was collected from the lowermost coarse pumice fall unit (Fig. 2, Loc. 2), because these fall units were heaped up during a short period of eruption.

#### 3. TL measurements

Sampling sites were covered with an opaque cloth. All samples were collected after the top surface layer of ca. 5 cm thickness was removed.

After PFG (ca. 4–10 µm) were separated by suspension in acetone, they were treated with 10% hydrogen peroxide for 16 hr and with 10% hydrochloric acid for 90 min. Then TL measurements were performed using a Daybreak TL reader (model 1150; Shitaoka and Nagatomo, 2011).

A standard MAAD protocol was used to determine palaeodoses. For each sample, three added doses (10, 20, and 30 Gy) were applied to sets of natural TL aliquots (generally five in each set). The equivalent dose  $(D_e)$  of the sample can be estimated by fitting the data assuming linear dose-dependence. Consequently, for correcting the nonlinear portion in the low-dose region (nonlinearity correction (Aitken, 1985; Shitaoka and Nagatomo, 2011),  $\Delta$  in Table 2), we measured the TL of the samples annealed at 350°C for 60 min and added irradiation at 20, 30, and 40 Gy.

The dose rate was estimated from uranium, thorium, and potassium concentrations measured using a high-purity Ge detector (EGSP 8785; Eurisys Mesures). The dose rates (alpha, beta and gamma) were calculated using the dose-rate conversion factors (Adamiec and Aitken, 1998). The dose rates were corrected using the present-day water contents (Zimmerman, 1971). Contributions of the cosmic dose rate to the dose rate were assumed as 0.15 mGy/yr by following Prescott and Hutton (1994) and Shitaoka *et al.* (2009a).

#### 4. Results and discussion

Fig. 3(a) presents TL measurement results. Minerals of PFG samples were mainly plagioclase, as inferred

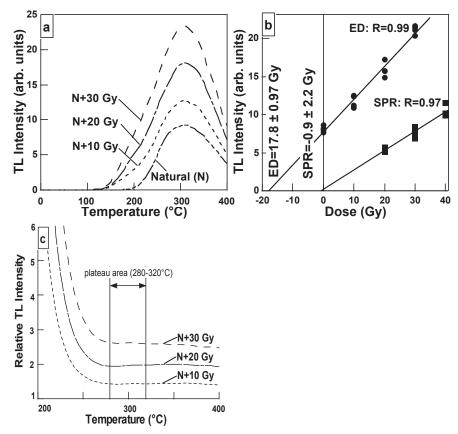


Fig. 3 TL glow curves (a) and TL growth curve (b) (Loc. 2). The equivalent dose (De) and the nonlinearity correction (SPR, Δ in Table 2) were estimated using the TL growth curve. The plateau area (Integrated area) is 280–320°C (c).

from TL signals shape and intensity, and from results of XRD analysis. The  $D_e$  of each sample can be converted to an apparent dose by extrapolation using a linear model of TL growth (see Fig. 3 (b)). The TL growth for each sample was obtained from each plateau area (Loc. 1 and Loc.2 were 310-330°C and 280-320°C (see Fig. 3 (c)), respectively), as determined using systematically performed plateau tests (Aitken, 1985). A lack of fading of TL signals within each plateau area was confirmed using plateau tests. The method of estimating nonlinearity correction ( $\Delta$  in Table 2) is analogous to that of  $D_e$  (Aitken, 1985; Shitaoka and Nagatomo, 2011). The TL growth for nonlinearity correction was obtained from each plateau area (e.g. 280-320°C). The palaeodose of the sample is the sum of the  $D_e$  and nonlinearity correction  $(\Delta)$ .

The dose rate was obtained as the sum of alpha, beta, gamma, and the cosmic dose rates. The palaeodoses, dose rates, and TL ages are presented in Table 2. The respective palaeodoses of Sz-S samples of Loc. 1

and Loc. 2 were 19.5  $\pm$  1.8 Gy and 16.9  $\pm$  2.4 Gy. The respective dose rates of Sz-S samples of Loc. 1 and Loc. 2 were 1.41  $\pm$  0.03 mGy/yr and 1.26  $\pm$  0.07 mGy/yr. We obtained the TL ages, 13.8  $\pm$  1.3 kyr in Loc. 1 and 13.4  $\pm$  2.0 kyr in Loc. 2. Uncertainties of each TL age were ca. 9–15%; they were mostly attributable to scattering of the glow curves of each aliquot and sensitivity changes of annealing samples for nonlinearity correction ( $\Delta$ ).

We calibrated the AMS- $^{14}$ C data (Okuno, 1997) tentatively in relation to the Sz-S bed using the IntCal13 dataset (Reimer *et al.*, 2013) (Table 1). Okuno (2002) obtained a calibrated age of 12,800 cal yrBP for Sz-S. The newly obtained TL ages in this study, 13.8  $\pm$  1.3 and 13.4  $\pm$  2.0 kyr, are fairly consistent with the seven calibrated- $^{14}$ C ages in this study (Table 1) within the error limits (Fig. 4).

Judging from the results and inferences presented above, age determination using TL method of PFG with the MAAD protocol is a powerful tool for quartz-less

Locality	Equivalent dose (Gy)	Δ (Gy)	Paleodose (Gy)	U (ppm)	Th (ppm)	K <sub>2</sub> O (wt%)	Water (wt%)
Loc. 1	17.1±1.2	2.4±1.3	19.5±1.8	1.04±0.08	7.16±0.36	1.04±0.07	81±1
Loc. 2	17.8±0.97	-0.9±2.2	16.9±2.4	1.05±0.14	7.32±1.29	0.85±0.10	79±1

Table 2 Results of TL dating of Sz-S

Alpha ray (mGy/yr)	Beta ray (mGy/yr)	Gamma+Cosmic ray (mGy/yr)	Dose rate (mGy/yr)	TL age (kyr)
0.37±0.01	0.53±0.03	0.51±0.01	1.41±0.03	13.8±1.3
0.32±0.04	0.45±0.04	0.48±0.04	1.26±0.07	13.4±2.0

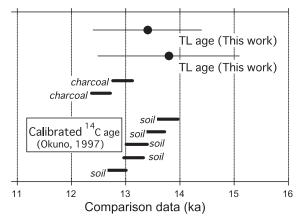


Fig. 4 TL age of Sz-S compared to calibrated <sup>14</sup>C age of samples related closely to the Sz-S layer.

tephra. However, the anomalous fading of PFG samples should be checked for each sample by plateau testing or fading testing.

### 5. Conclusion

We measured TL ages using PFG at MAAD protocol for Sz-S tephra of Sakurajima volcano to confirm estimates of their ages based mainly on the  $^{14}$ C dates. The TL ages obtained from the tephra itself were 13.8  $\pm$  1.3 and 13.4  $\pm$  2.0 kyr, which are fairly consistent with the calibrated age for those  $^{14}$ C dates of charcoal and soil samples closely related to the tephra layer.

Results show that TL ages obtained from the present study are useful for accurate estimation of the Sz-S eruption age.

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#### References

Adamiec, G. and Aitken, M.J. (1998) Dose-rate conversion factors: update. Ancient TL, 16, 37-50.

Aitken, M.J. (1985) Thermoluminescence Dating. Academic Press, 359pp.

Aramaki, S. (1984) Formation of the Aira Caldera, Southern Kyushu, ~22,000 years ago. Journal of Geophysical Research, 89, 8485-8501.

Buvit, I., Izuho, M., Terry, K., Shitaoka, Y., Soda, T. and Kunikita, D. (2014) Late Pleistocene Geology and Paleolithic Archaeology of the Shimaki Site, Hokkaido, Japan. Geoarchaeology, 29, 221-237.

Ganzawa, Y., Usui, R., Tanaka, H. and Azuma, T. (2007) Red thermoluminescence dating of Toya pyroclastic flow using single-aliquot regenerative-dose (SAR) protocol. Jour. Geol. Soc. Japan, 113, 470-478. (in Japanese, with English Abstract)

Gillot, P.Y., Valladas, G., and Reyss, J.L. (1978) Dating of lava flow using a granitic enclave: Application to the Laschamp magnetic event. PACT (Journal of the European Study

- Group on Physical, Chemical and Mathematical Techniques Applied to Archaeology), 2, 165-173.
- Guerin, G. and Valladas, G. (1980) Thermoluminescence dating of volcanic plagioclases. Nature, 286, 697-699.
- Ichikawa, Y., Hagihara, N., and Nagatomo, T. (1982) Dating of pyroclastic flow deposits using the quartz inclusion method. PACT (Journal of the European Study Group on Physical, Chemical and Mathematical Techniques Applied to Archaeology), 6, 409-416.
- Kobayashi, T. (1986) Volcanic history and pyroclastic flows of Sakurajima Volcano. In: Aramaki, S. (Ed.), Characteristics of dry high concentration flows (Pyroclastic Flows) associated with volcanic eruption and their disasters, 137-163. (in Japanese)
- Kobayashi, T. and Fukushima, H. (1984) Tephrochronology of Sakurajima volcano. Bulletin of the volcanological society of Japan, 29, 322. (in Japanese)
- Kobayashi, T. and Ezaki, M. (1997) Review of Eruptive history of Sakurajima volcano. Monthly Chikyu, 19, 227-231. (in Japanese)
- Kobayashi, T. and Tameike, T. (2002) History of eruptions and volcanic damage from Sakurajima Volcano, southern Kyushu, Japan. The Quaternary Research of Japan, 41, 269-278. (in Japanese, with English Abstract)
- Kodama, K. (2001) From the Paleolithic to the Jomon period in southern Kyushu, Japan. The Quaternary Research, 40, 499-507. (in Japanese)
- Lowe, D.J. (2011) Tephrochronology and its application: a review. Quaternary Geochronology, 6, 107-153.
- Machida, H. and Arai, F. (1992) Atlas of Tephra in and around Japan. The University of Tokyo Press, Tokyo. (in Japanese)
- Murray, A.S. and Wintle, A.G. (2000) Luminescence dating of quartz using an improved single-aliquot regenerative-dose protocol. Radiation Measurements, 32. 57-73.
- Moriwaki, H. (1992) Late quaternary phreatomagmatic tephra layers and their relation to paleo-sea levels in the Aira caldera, southern Kyushu, Japan. Quaternary International, 13/14, 195-200.
- Moriwaki, H. (1994) Palaeoenvironmental study of Late Quaternary Fine Ash Beds around Kagoshima Bay. Reports for Grants-in-Aid for Scientific Research (no. 04680248) from Ministry of Education. (in Japanese)
- Okuno, M. (1997) Radiocarbon chronology of Sakurajimatephra group. Monthly Chikyu, 19, 231-235. (in Japanese)
- Okuno, M. (2002) Chronology of tephra layers in southern Kyushu, SW Japan, for the last 30,000 years. The Quater-

- nary Research of Japan, 41, 225-236. (in Japanese, with English Abstract)
- Oishi, M., Geshi, N. and Shitaoka, Y. (2011) Identification of volcanic products around the southern foots of Haruna volcano, central Japan: an example of identifying volcanic products using refractive indices of plagioclase phenocrysts. The quaternary research, 50, 295-308. (in Japanese, with English Abstract)
- Prescott, J. R. and Hutton, J. T. (1994) Cosmic ray contributions to dose rates for luminescence and ESR dating: Large depths and long-term time variations. Radiation Measurements, 23, 497-500.
- Reimer, P.J., Bard, E., Bayliss, A., Beck, J.W., Blackwell, P.G., Ramsey, C.B., Buck, C.E., Cheng, H., Edwards, R.L., Friedrich, M., Grootes, P.M., Guilderson, T.P., Haflidason, H., Hajdas, I., Hatté, C., Heaton, T.J., Hoffman, D.L., Hogg, A.G., Hughen, K.A., Kaiser, K.F., Kromer, B., Manning, S.W., Niu, M., Reimer, R.W., Richards, D.A., Scott, E.M., Southon, J.R., Staff, R.A., Turney, C.S.M. and van der Plicht, J. (2013) Intcal 13 and Marine 13 radiocarbon age calibration curves 0–50,000 years cal BP. Radiocarbon, 55, 1869-1887.
- Shitaoka, Y. and Nagatomo, T. (2011) Age determination of maker tephras relative to paleolithic sites based on luminescence dating for reading natural site formation process: a case study of paleolithic sites in southern Miyagi. Archaeology and natural science, 62, 73-84. (in Japanese, with English Abstract)
- Shitaoka, Y., Nagatomo, T. and Obata, N. (2009a) Age determination of Ontake Pm1 pumice fall deposit (On-Pm1) by thermoluminescence method. The Quaternary Research, 48, 295-300. (in Japanese)
- Shitaoka, Y., Fukuoka, T., Hasegawa, A., Kusano, T. and Nagatomo, T. (2009b) Thermoluminescence dating of the pyroclastic deposits of Sanbe Volcano. Bulletin of the Shimane nature museum of Mt. Sanbe (Sahimel), 7, 15-24. (in Japanese, with English Abstract)
- Toyoda, S., Tsukamoto, S., Hameau, S. Usui, H. and Suzuki, T. (2006) Dating of Japanese quaternary tephras by ESR and luminescence methods. Quaternary geochronology, 1, 320-326.
- Wintle, A.G. (1973) Anomalous Fading of Thermo-luminescence in Mineral Samples. Nature, 245, 143-144.
- Zimmerman, D.W. (1971) Thermoluminescence dating using fine grains from pottery. Archaeometry, 13, 29-52.

# 熱ルミネッセンス年代測定による桜島薩摩テフラの噴火年代

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#### 要 旨:

桜島火山起源である桜島薩摩テフラ(Sz-S)の噴火年代を、熱ルミネッセンス(TL)年代測定法を用いて年代測定を行った。Sz-S は、2カ所の露頭で試料採取した。試料処理は多鉱物微粒子法(PFG)を用い、付加線量法で TL 測定を行った。得られた TL 年代は、 $13.8\pm1.3$  kyr と $13.4\pm2.0$  kyr であった。Sz-S 中から採取された木炭や Sz-S 直下の土壌から得られた放射性炭素( $^{14}$ C)年代(奥野、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を、1997)を用いて暦年較正した 19970年代と今回得られた TL 年代を比較したところ、誤差の範囲で整合性のある結果であった。よって、19970年代とされる Sz-S の噴火年代は、約13 kyr であり、既存の 19970年代データと矛盾がないことがわかった。

キーワード: 桜島薩摩テフラ、新期北岳段階、熱ルミネッセンス年代測定、放射性炭素年代測定、噴火年代